

**APPLICATIONS OF SQUEEZED STATES: BOGOLIUBOV
TRANSFORMATIONS AND WAVELETS TO THE
STATISTICAL MECHANICS OF WATER AND ITS BUBBLES**

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ABSTRACT

The *squeezed states* or *Bogoliubov transformations* and wavelets are applied to two problems in non-relativistic statistical mechanics: the dielectric response of liquid water, $\epsilon(\vec{q}, \omega)$, and the bubble formation in water during insonnification. The *wavelets* are special phase-space windows which cover the domain and range of $L^1 \cap L^2$ of classical causal, finite energy solutions. The multi-resolution of discrete wavelets in phase space gives a decompostion into regions of time and scales of frequency thereby allowing the renormalization group to be applied to new systems in addition to the tired 'usual suspects' of the Ising models and lattice gasses. The Bogoliubov transformation: squeeze transformation is applied to the *dipolaron* collective mode in water and to the gas produced by the explosive cavitation process in bubble formation.

1. INTRODUCTION

Water is extremely important in chemistry and biology both as a solvent and as a neutral medium which is rich in possibilities.¹⁻¹⁹ The dielectric response of a medium, $\epsilon(\vec{q}, \omega)$ is related to the index of refraction as, $n(\vec{q}, \omega) = \sqrt{\epsilon\mu}$, whose real part controls wave propagation and whose imaginary part gives the attenuation. An August 1993 dissertation at Missouri University by one of us, S.-H. Kim¹⁹, formulates a new model which is rather successful in accounting for the complex index of refraction $n(\omega) + i\alpha(\omega)$, over 14 to 15 decades of frequency. This model is a number-conserving relaxation-time,⁷ Boltzmann transport equation model in phase space^{5,6,9} based on Kubo's fluctuation-dissipation theorem¹² and a Kerr approximation.¹¹ The free rotation gives an IR peak at the correct frequency which is much larger than the one found experimentally by Simpson et al.¹⁵ A phase-space analysis of our model based on wavelets will be presented here.

The problem of bubble formation in liquids is both old and difficult.²⁰⁻²⁵ It is now known that two different mechanisms of bubble formation exist: **cavitation**, where the growth is explosive and **nucleation** where diffusion of molecules yields a slow growth. This work will be restricted to cavitation. All bubbles are interesting for electromagnetic wave propagation in water because they give additional spatial dispersion \vec{q} -dependence to the index of refraction of the medium through their Mie scattering.

The approach taken here will be to form a mean-field theory of a Φ^4 theory in 4 space-time dimensions. It is a variant of an idea due to Kaup,²⁵ Wilhelmsson²⁶ and Glimm²⁷ that bubbles are non-linear, coherent effects in fluids. Kaup's clever title is, "Cavitons are solitons" for a plasma driven by both sound and electromagnetism. Our equations are completely different and have solitary wave solutions rather than solitons. These solitary waves will have finite lifetimes in contrast to the infinite lifetimes of Kaup's cavitons, which

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he first showed were an integrable system. However, since both models are subject to other perturbations, this difference is not actually as serious as it first appears.²⁹

There is also a practical reason for trying to understand bubble formation. Apfel^{30,31}, Nyborg³², Rose and Goldberg³³ have shown that ultrasound at the frequencies used for medical diagnostics forms bubbles in water and tissue. They have raised questions about the safety of ultrasound but have not yet shown any evidence that these bubbles (or any other effect of ultrasound) are harmful. Trevena³⁴ has suggested that bubble collapse may occur through shock waves, which could damage cells.

Both of these problems will be analyzed using wavelets³⁶⁻⁵¹ which are a special class of **windows**

- (i) which cover the domain \mathbb{R}^1 or \mathbb{R}^2 in refs. (36-49) and the Lorentzian manifold of special relativity in ref (50,51); and
- (ii) are dense in the range of $L^2 \cap L^1$.

The first space L^2 is required classically for finite energy signals and for a probability interpretation of Hilbert space states in non-relativistic quantum mechanics, and the space L^1 is required classically for causality and is a major technical requirement in non-relativistic quantum mechanics which provides absolute continuity. Clearly, (i) assures the existence of measurability of the underlying space and (ii) places additional restrictions on the solutions for the correct dynamics and symmetries. In particular, the symmetry group is the set of **dilations**. In the continuous one-dimensional case dilations are generated by the transformation D

$$(D_{ab})(x) = ax + b \quad (1)$$

$$(D_{ab}\psi)(x) = |a|^{1/2}\psi(ax - b) \quad (2)$$

on the coordinates x and solutions $\psi \in L^2 \cap L^1$, where $a \in \mathbb{R}_+^1$ and $b \in \mathbb{R}^1$. In the discrete case $(k, j) \in$ (the integers) with $a = 2$ (one choice, but not the only one)

$$(D_{jk})(x) = 2^j \cdot x + k \quad (3)$$

$$(D_{jk}\psi)(x) = \psi_{jk}(x) = |2|^{j/2}\psi(2^j x - k) \quad . \quad (4)$$

In the discrete case, $L^2 \rightarrow \ell^2$ and $L^1 \rightarrow \ell^1$ and ψ_{jk} "live in" $\ell^2 \cap \ell^1$. The choice of $a = 2$ gives the Lebeque covering of \mathbb{R}^1 and extends to \mathbb{R}^n and thus any other integer with this property will suffice. To satisfy (2) the conditions (2a) completeness

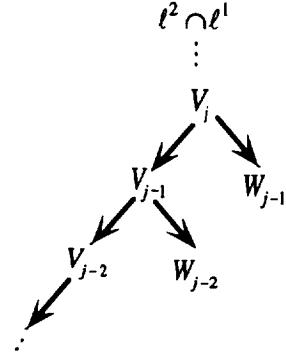
$$\overline{V\psi_{jk}} = L^2 \cap L^1 \quad (5)$$

(2b) **mean zero** for ψ , mean one for ϕ

$$\hat{\psi}(0) = \int_{-\infty}^{\infty} \psi(x)dx = 0 \quad (6)$$

$$\hat{\phi}(0) = \int_{-\infty}^{\infty} \phi(x)dx = 1 \quad (7)$$

where the multiresolution decomposition of Mallet satisfies $V_j = V_{j-1} \overline{\oplus} W_{j-1}$, i.e. $V_{j-1} \cup W_{j-1} = V_j$ and $V_{j-1} \cap W_{j-1} = \{0\}$. For fixed j , eqs. (3,4) reduce to a translation on ℓ^2 .



If k is considered as a time (space) translation, kt_0 , and 2^j as a frequency (wavenumber) scale, $2^j\omega_0$, with $t_0\omega_0 \geq 1$ then each fixed j_0 corresponds to a discrete translation group at frequency scale $2^{j_0}\omega_0$ and changing j_0 changes the frequency scale. Even if ψ or ϕ are time (space) signals, the dilation operators D_{ab} or D_{jk} map them into the time (space)—frequency (wave number) **phase space**. Thus

$$\phi(x) \in V_j \quad \text{and} \quad \psi(x) \in W_j \quad (8)$$

implies that

$$\phi(2x) \in V_{j-1} \quad \text{and} \quad \psi(2x) \in W_{j-1} \quad . \quad (9)$$

The interpretation of the V_j 's spanned by the ϕ_j 's is as a low band-pass window at frequency scale $2^j\omega_0$ whereas the higher frequencies are in the W_j 's spanned by ψ_j 's at that frequency scale.

From this short discussion two things should be obvious. One is that although a wavelet is a window, it is a great deal more as it must cover each space-time point in order not to miss any point particles, or sources of charge or mass. The other is that all of the solutions allowed by the symmetries and dynamics of the system must be expressed in the wavelet basis, i.e. they must be a complete set. Kaiser⁵⁰ has formulated relativistic electrodynamics in terms of a wavelet based on analyticity and coherent states. D'Aranio and DeFacio⁴⁹ have formulated a class of squeezed states of quantum optics and provided a general inversion structure for a general density operator in terms of a suitable window, which was expressed in terms of a general basis of observables. Han, Kim and Noz⁵¹ have presented a study of the relativistic phase space of light using compactly supported wavelet windows to clarify the relation between photons and light waves. Since there are many wavelet windows for any given problem, it is important to know which ones are natural and, if possible, if a "best one" exists.

In this paper, the application of wavelets and state squeezing, Bogoliubov transformations to the two statistical mechanics problems: $\in (\vec{q}, \omega)$ for water in **Section 2** and bubble formation will be described in **Section 3**, and our conclusions will be presented in **Section 4**.

2. STATISTICAL MECHANICS OF WATER, THE DIELECTRIC RESPONSE

In collaboration with Professor G. Vignale, we have published studies of a classical water-like polar fluid whose properties are chosen to mimic water. The starting point is the number-conserving relaxation-time approximation to the Boltzmann transport equation. Up to the first order deviation from equilibrium it has the form

$$\frac{\partial f_1}{\partial t} + \sum_{\alpha} \left[\dot{q}_{\alpha} \left(\frac{\partial f_1}{\partial q_{\alpha}} \right) + \dot{p}_{\alpha} \left(\frac{\partial f_1}{\partial p_{\alpha}} \right) - \left(\frac{\partial v}{\partial q_{\alpha}} \right) \left(\frac{\partial f_0}{\partial p_{\alpha}} \right) \right] = -\frac{f_1 - f_1^{loc}}{\tau} , \quad (10)$$

where overdots are time rate of changes, $\dot{q}_{\alpha} = \frac{\partial T}{\partial p_{\alpha}}$, $\dot{p}_{\alpha} = -\frac{\partial T}{\partial q_{\alpha}}$, $f_0 = e^{-\beta T}/Z$ is the equilibrium Boltzmann distribution function. T is the kinetic energy of the molecule and includes translations, rotations and small amplitude vibrations, and Z is the partition function. f_1 is the first order correction term to f_0 by a perturbed applied potential. The static response of the electric susceptibility $\chi(\omega = 0)$ is a 3×3 real matrix-valued quantity given by $-\beta f_0(q_{\alpha})$.

Upon determining the dynamical self-response function $\chi_s(\omega)$ self-consistently from f_1^{loc}

$$\chi_s(\omega) = (1 + i/\omega\tau) \chi^0(\omega + i/\tau) \left[1 + \frac{i}{\omega\tau} \chi^0(\omega + i/\tau) \{ \chi^0(\omega = 0) \}^{-1} \right]^{-1} , \quad (11)$$

where the Kubo fluctuation dissipation theorem for the single particle self-electric susceptibility

$$\chi^0(\omega) = \chi(0) [1 + i\omega G(\omega)] , \quad (12)$$

relates the electric susceptibility to the van Hove correlation function $G(\omega)$. The total response from the Kubo theorem is

$$\chi(\omega) = \chi_s(\omega) [1 - \Psi(\vec{q}) \chi_s(\omega)]^{-1} , \quad (13)$$

where Ψ is the local field factor which describes the coherent many particle interactions. It is the analog of Lorentz's local field factor, in modern many-body theory. We have explored several choices and found Wei and Patey¹⁷ to provide the best agreement with experiment. Indeed, various computer intensive molecular dynamics and Monte Carlo calculations gave $\epsilon_T(0, 0)$ as 25-71 instead of the experimental value of 90.8 ± 3.2 . The large experimental uncertainty is dominated by the spontaneous dissociation $H_2O \rightleftharpoons H^+ + OH^-$, which at $20^\circ C$ yields an ion concentration of 8.33×10^{-8} moles/liter. The high mobility of the H^+ species allows it to form a positively charged surface layer at the negative electrode which can distort the measured value of the electric permittivity, $\epsilon(\omega)$. By taking a limit as the moments of inertia approach zero, the Stockmayer fluid calculations were found comparable to the best computer studies. The causal dielectric permittivity values are

$$\frac{1}{\epsilon_T(\vec{q}, \omega)} = \frac{1}{[1 + 4\pi\chi_T(\vec{q}, \omega)]} \quad (14a)$$

and

$$\frac{1}{\epsilon_L(\vec{q}, \omega)} = [1 - 4\pi\chi_L(\vec{q}, \omega)] \quad (14b)$$

where (T, L) denote the transverse and longitudinal components of the dielectric tensor ϵ and the electric susceptibility tensor χ . The directions (T, L) are taken from the direction of the applied (vacuum) electric field \vec{E} . **Remark:** A wave propagation or scattering experiment is required to measure the \vec{q} dependence of n and α , in contrast to the *ac* capacitance bridge which measures only the angular frequency, ω , dependence. A graph from ref. (18) of the real part of the index of refraction will be shown later.

There is a collective mode in ϵ_L which Lobo, Robinson and Rodrigues⁹ first suggested and Pollock and Alder¹⁰ found in their simulation of a Stockmayer fluid model of water. It occurs in our model both in the symmetric rotor “waterlike” case¹⁸ and in the realistic asymmetric rotor case.¹⁹

Considering the water molecule as a two-level system for simplicity, the composite Boson creation and destruction operators which create and destroy water molecules (*recall* that this analysis is at frequencies that are far below the breakup threshold energies) are written as $(\vec{a}_\alpha, \vec{a}_\beta^\dagger)$ where their directions are those of the electric dipole moment, $\vec{\mu}$, of the molecule and $\alpha, \beta = 1, 2$ label which level. The collective dipolaron boson is created and destroyed by the Bogoliubov transformed operators $(b_\alpha, b_\beta^\dagger)$. The frequency of the dipolaron is complex-valued

$$\omega_\alpha = \Omega_{q\alpha} - i\Gamma_{q\alpha} \quad (15)$$

and lies in the lower half ω -plane because the response function $1/\epsilon_L(\vec{q}, \omega)$ is causal. The frequency in eq. (15) is determined from

$$Re [\epsilon_L(\vec{q}, \Omega_{q\alpha})] = 0 \quad , \quad (16)$$

and the width from

$$\Gamma_{q\alpha} = \frac{|Im [\epsilon_L(\vec{q}, \Omega_q)]|}{\left| \left[\frac{\partial \epsilon_L(\vec{q}, \omega)}{\partial \omega} \right] \right|_{\omega=\Omega_{q\alpha}}} \quad , \quad (17)$$

provided that $\Omega_{q\alpha} \gg \Gamma_{q\alpha}$. The peak and its width are shown in Fig. 1, where one can see that $\Omega_{q\alpha} \gg \Gamma_{q\alpha}$ is satisfied. The mechanism of this collective mode is high frequency coherent oscillations of the electric dipoles in self-consistent fields in the liquid. In order for the mode to be longitudinal, they oscillate 180° out of phase. Both $\Omega_{q\alpha}$ and $\Gamma_{q\alpha}$ were shown by Kim, *et al.*¹⁸, to be almost *dispersion-free* (they only change by 1% over the entire range of validity). This mode depends on $q = |\vec{q}|$ being small but non-zero so it cannot be measured from any *ac* capacitance experiment such as those in ref. (3) and must await a light or neutron scattering experiment. Such an experiment would *verify* or *falsify* the model formulated in refs. (18,19).

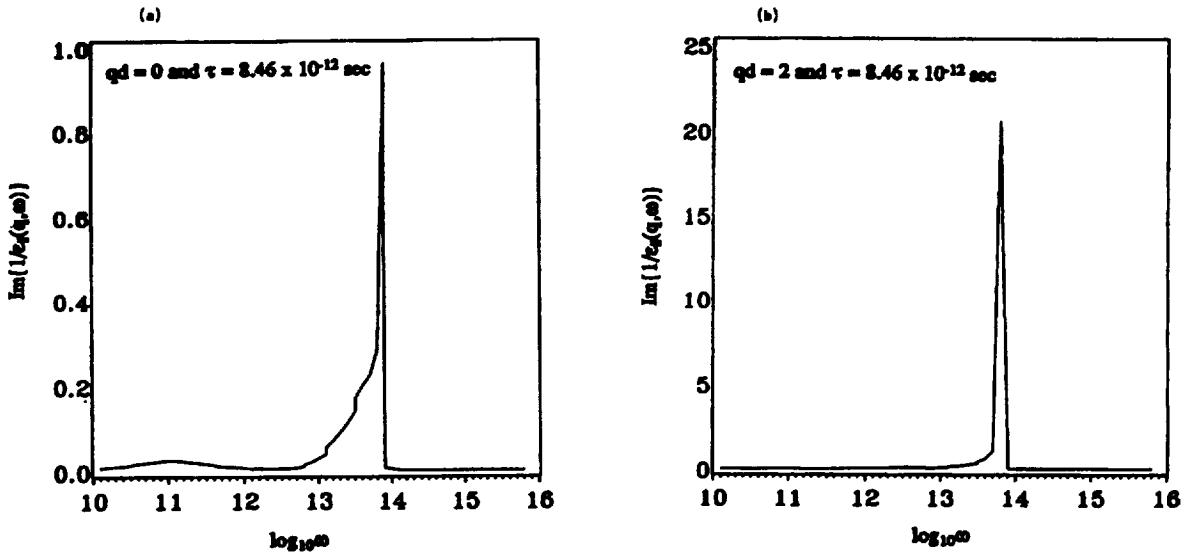


Fig. 1. The dipolaron collective mode in water.

Next, the reader is reminded that eq. (10) includes translations, rotations and vibrations in the $\dot{p}_\alpha \left(\frac{\partial T}{\partial p_\alpha} \right)$ terms. It is well known^{52,53} that the renormalization group is based upon a scaling invariance. By expanding the single particle distribution function $f_1(\cdot)$ in a wavelet series

$$f_1(\cdot) = \sum_{jk} C_{jk} \psi_{jk}(\cdot) \quad (18)$$

where (\cdot) can stand for either the translation \vec{x} , rotation (θ, ϕ) or vibrations (q_1, q_2, q_3) in the real index of refraction $n(\omega)$. The three normal modes for vibration are given by group theory and are used in our calculation of $n(\omega)$. In Fig 2 the frequencies up to $10^{10} Hz$ are dominated by the translations, from $10^{10} \sim 10^{12} Hz$ at (A, A') the collisions dominated Debye relaxation time dominates and at $10^{13} Hz$ the free rotor peak occurs at (B_1, B') . The free rotor peak at (B') in our theory is much too large, so work is underway to try to bring this into better agreement with the experiment. Between 10^{13} and $10^{14} Hz$ the collective mode (C') occurs with coherent oscillations of the dipoles as its mechanism. The small optical peaks at (D_1, D_2) fail to resolve the two higher frequency modes in (D_2) but we think that future experiments will. Thus, the wavelet phase space allows us to analyze the Boltzmann transport equation with a non-trivial interaction.

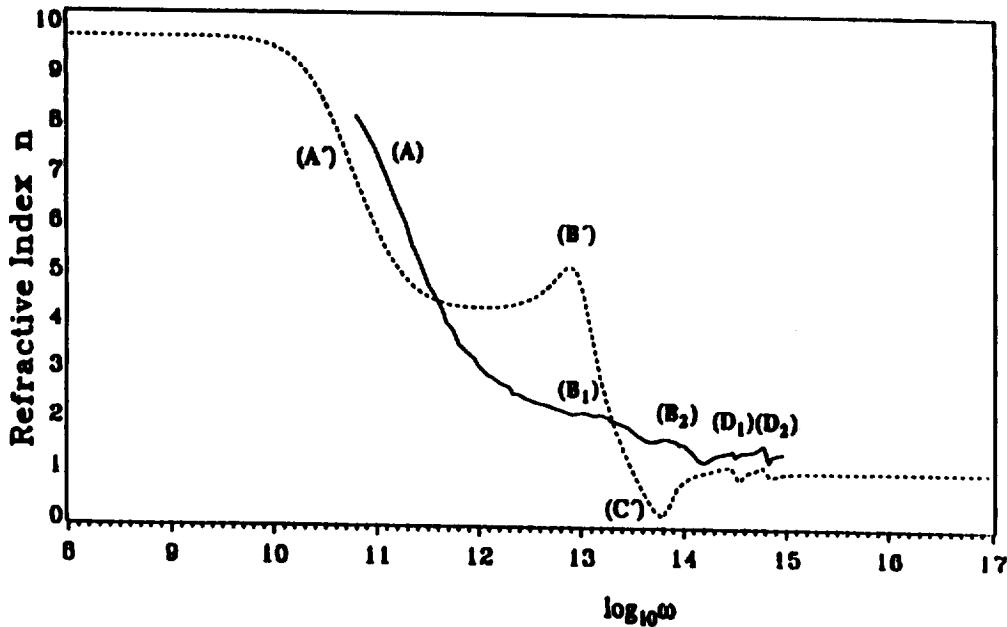


Fig. 2. The real index of refraction of water.

3. BUBBLE FORMATION IN WATER BEING INSONIFIED BY ULTRASOUND

The idea behind this calculation is to adapt the idea of Kaup to a solitary wave model by generalizing the work of Hammer, Shrauner and DeFacio where ϕ is a $1 \oplus 1$ fluid density field

$$L = -\frac{1}{2} [(\phi_x)^2 + (\phi_t)^2] - \frac{B}{4} (\phi^2 - u_0^2)^2 , \quad (19)$$

with $u_0^2 = A/B$ where A, B are real and positive coupling constants and A is the strength of the negative mass, B is the strength of the positive nonlinear quartic interaction to three space dimensions with a radial time independent potential. In $3 \oplus 1$ space-time dimensions

$$L_F = -\frac{1}{2} [(\vec{\nabla} \phi)^2 + \phi_t^2] - \frac{B}{4} (\phi^2 - u_0^2)^2 , \quad (20a)$$

$$L_A = -\frac{1}{2} [(\vec{\nabla} p)^2 - p_t^2] - \frac{B_1}{4} (p^2 - u_{01}^2)^2 , \quad (20b)$$

$$L_I = g\phi^2 p^2 + \mu\phi_t^2 \quad (20c)$$

where L_F is the Lagrangian of the fluid, L_A is the acoustic Lagrangian for the sound wave and L_I is the interaction Lagrangian. If $\mu \approx 0$ and the sound waves are small enough in amplitude to be linear (see eq. (30a)) the field equations become

$$-(\Delta\phi + \phi_{tt}) - A\phi + B\phi^3 + gp^2\phi = 0 , \quad (21a)$$

$$-(\Delta p - p_{tt}) - A_1p + B_1\rho^3 + g\phi^2p = 0 \quad (21b)$$

The key role played by the nonlinear "potential" which acts as a *convection* term is our justification for calling this approach a *strong convection model*. The mechanism is that

the constant sound wave in the fluid produces gas under pressure as a new *phase* in the fluid. This is a non-equilibrium, open system and the gas density can be described by one Bogoliubov transformation and the non-zero temperature, 0°C to 100°C , can be obtained from another Bogoliubov transformation. The entropy of this situation was discussed by DF, VN and Professor Brander⁶³ of the Institute for Theoretical Physics at Chalmers in the Göthenberg. The $\ell = 0$, radial solutions to eqs. (21a,b) are Jacobi elliptic functions with modulus k_1 , $0 \leq k_1 \leq 1$, δ_{01} is a density defect where the local mass density is a minimum so that it is a site for cavitation and (u, u_{01}) are the amplitudes for the density and pressure wave, respectively.

Next a time-independent *mean field theory* for the density ϕ is formed by linearizing about the Jacobi elliptic functions according to

$$gp^2\phi \rightarrow gu_{01}^2 [Sn(\kappa_1(r + \delta_{01})|k_1)]^2 \phi \quad , \quad (22)$$

and

$$B\phi^3 \rightarrow B <\phi^2> \phi \rightarrow Bu^2 [Sn(\kappa(r + \delta)|k)]^2 \quad . \quad (23)$$

This should be valid, provided it is not applied too close to a caviton ‘explosion’, and leads to the linearized field equation for the density ϕ

$$\Delta\phi + \phi_{tt} + V_{MFT}(r)\phi = 0 \quad , \quad (24)$$

where

$$V_{MFT}(r) = A - Bu^2 [Sn(\kappa(r + \delta)|k)]^2 + gu_{01}^2 [Sn(\kappa_1(r + \delta_{01})|k_1)]^2 \quad (25)$$

Separation of variables using

$$\phi = R(r)Y_{\ell m}(\theta, \Phi)e^{-\omega_q t} \quad , \quad (25a)$$

and

$$R(r) = \frac{u(r)}{r} \quad (25b)$$

reduce eq. (25) to

$$u_{rr} + \left[V_{MFT} - \frac{\ell(\ell t^1)}{r^2} + \omega_q^2 \right] u = 0 \quad . \quad (26)$$

This ODE satisfies Dirichlet zero boundary conditions on the dense domain $H^2(R_+^1)$ where R_+^1 is the positive half-line and H^2 is the Sobolev space of L^2 functions whose first two derivatives exist and are L^2 . Thus, eq. (26) can be solved numerically using the Numerov algorithm. Then the density is constructed from the relation

$$\phi(\vec{r}, t) = \sum_{q\ell m} c_{q\ell m} R_{q\ell}(r) Y_{\ell m}(\theta, \phi) e^{-\omega_q t} \quad . \quad (27)$$

From eq. (27) the sound wave has modulus $k_1 \simeq 0$. One thing about this strong convection model which seems new is the inclusion of $\ell \geq 1$ multipoles which give deviations from spherical symmetry. The calculus of variation “proof” that bubbles are spherically shaped

only holds for thermal and fluid equilibrium, whereas the continuous quasi-monochromatic sound wave makes this an open, non-equilibrium system. In Fig. 3 V_{MFT} is plotted and

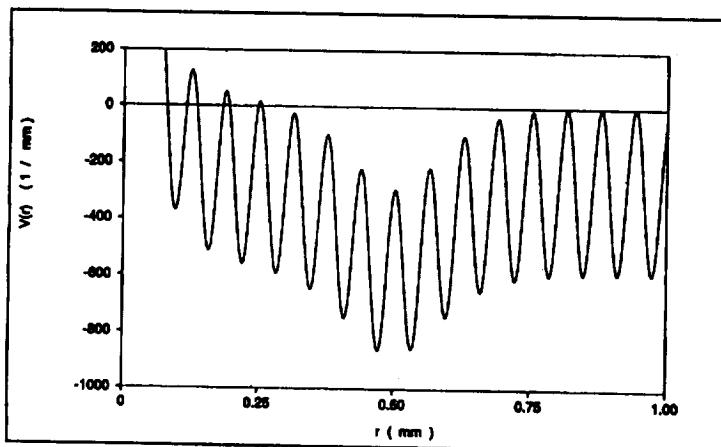


Fig. 3. An example of a mean field theory potential, V_{MFT} .

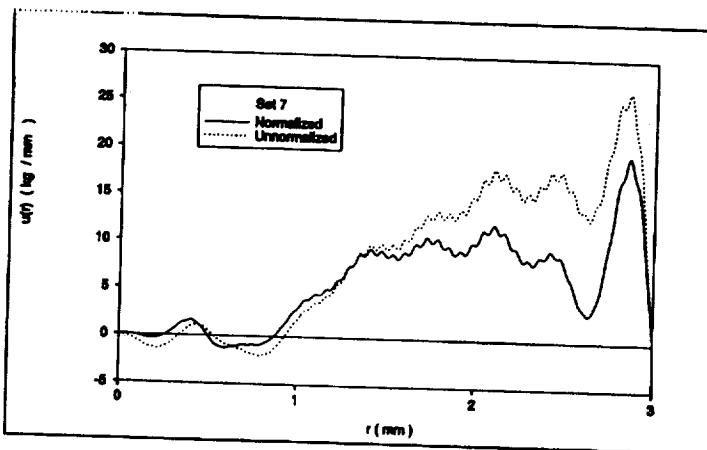


Fig. 4. The water density in a 3mm cell calculated from the mean field theory potential in the previous figure.

in Fig. 4 a bubble is shown which was found in the density, using the convection potential of Fig. 3. The parameters are consistent with those of water with a sound wave amplitude which is physically realizable. The bubble is centered at $\delta = 0.75\text{mm}$; and has radius $r_0 \simeq 300\mu$ ($\mu = 10^{-6}\text{m}$). The detection of bubble interfaces using wavelets is under study. Since some authors have questioned the safety of ultrasound in tissue,^{31,34} it is important to understand bubble formation. In addition, the Mie scattering from bubbles adds spatial dispersion to the effective index of refraction.

4. CONCLUSIONS

Two different problems in non-equilibrium statistical mechanics were discussed:

- (i) the dielectric response of water, $\epsilon(\vec{q}, \omega)$, and
- (ii) bubble formation in water under ionization using Bogoliubov transformations: state squeezing maps and wavelets.

The multi-resolution structure of phase space permits the sort of analysis which the renormalization group has provided for Ising models and lattice gas models and little else. Thus, "squeezed states" are useful for more than quantum optics.

ACKNOWLEDGEMENTS

This work was supported in part by the Missouri University Department of Physics and AFOSR grants 90-307 and 91-203. Professors Giovanni Vignale and Peter Pfeifer are thanked for many useful conversations. The figures were produced by the NeXT and Silicon Graphics Personal Iris work stations purchased by the two AFOSR grants. The organizers of the Third International Workshop on Squeezed States, Professors M.H. Rubin and Y.-H. Shih of UMBC and Dr. D. Han of NASA, Greenbelt are thanked for organizing an excellent conference which continues the traditions initiated by Professor Y.S. Kim and Professor W.W. Zachary.

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